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Reactive transport modeling of contaminant release from encased unirradiated fuel to Hanford sediments

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Unirradiated nuclear reactor fuel, colloquially named the uranium billets, was disposed via grout encapsulation in a designated trench at the Hanford Site located in the southeastern portion of Washington state, USA. The uranium billets were placed in boxes that were grout filled and encased in a series of concrete encasement forms resulting in a uranium billet monolith. The uranium billets have an approximate total inventory of 824 metric tons of uranium (roughly 687 Ci) with entrained technetium-99 concentrations ranging from 1 to 10 ppm from impurities due to waste recycling and recovery operations.

To evaluate the long-term impacts on the environment, a reactive transport model was developed to simulate geochemical reactions and transport of U-238 and Tc-99 out of the monolith by coupling GoldSim Pro with PhreeqcRM (an open source U.S. Geological Survey software) via dynamic-link library (DLL). Three major components are defined for the model including the grout-filled uranium billet waste form, surrounding encasement concrete, and stabilizing soil or backfill. A scaled conceptualization of the uranium billet monolith is developed in GoldSim Pro with diffusive transport as the dominant transport mechanism from the waste form to the surrounding soil. Radionuclides that are released from the uranium billet waste form transport through the encasement concrete and release into the surrounding stabilizing soil or backfill. A DLL interface between PhreeqcRM and GoldSim Pro has been developed to provide the capability to perform a variety of low-temperature aqueous geochemical calculations coupled with the transport model.

A simplified geochemical model is applied to each of the major model components defining both the mineral phases and starting solution composition. With each timestep, contaminant transport is calculated by GoldSim Pro with results passed to PhreeqcRM to perform geochemical calculations and solve for the evolving geochemical conditions. Model results indicate $Na_2U_2O_7$ and soddyite precipitate in the encasement concrete and soil, respectively, controlling U-238 release via solubility limited concentration gradient. Empirically derived data were compared to model results to evaluate model confidence and representativeness. Sensitivity and uncertainty analyses were performed to understand model behavior.